Hydrogen production process involves recycle loop in which tail gas produced downstream in the process is used upstream of the process at shift reactor, providing heat for drying coal and providing heat for generating carbon dioxide

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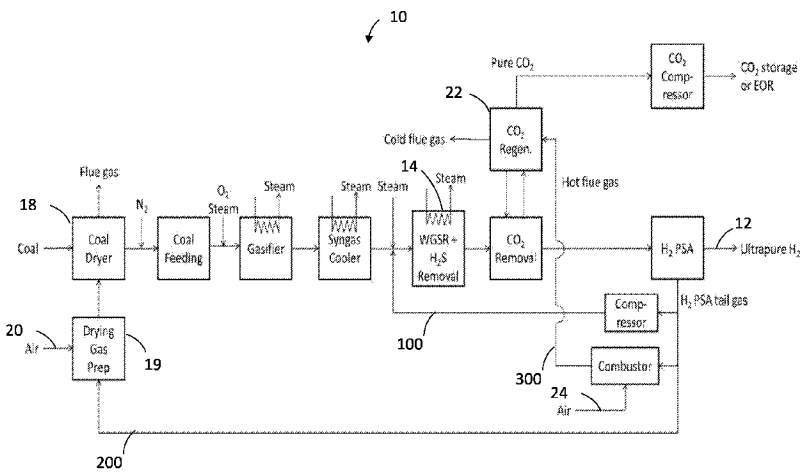

Figure 2

(57) **Abstract**: A hydrogen production process and production plant (10) operable to produce, at least, Hydrogen and Carbon dioxide, the process comprises a recycle loop, wherein tail gas (100, 200, 300) produced downstream in the process is used upstream of the process at a shift reactor and at least one of providing heat for drying coal (18) and providing heat for regenerating CO_2 (22).

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HYDROGEN PRODUCTION PROCESSING

FIELD OF INVENTION

The present invention relates to a hydrogen production process and a hydrogen production plant which utilises downstream tail gas for use at one or more upstream stages in the process to improve hydrogen production rate and reduce energy consumption involved.

BACKGROUND TO THE INVENTION

Gasification is the conversion of an organically derived, carbonaceous material by partial oxidation into a gaseous product, synthesis gas ("syngas") comprising hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), Nitrogen (N_2) and other hydrocarbons and impurities. Reactions are generally carried out at elevated temperatures and atmospheric or elevated pressures.

Hydrogen Pressure Swing Adsorption (H_2 PSA) is an example of a gas purification process, which is considered unique in the field of producing ultrapure hydrogen. For example a system having two to twenty columns per one train and having each column interconnected can produce an ultrapure hydrogen product continuously with high product recovery and productivity. The system is considered capable of producing/outputting a very high purity of hydrogen suitable for use as a refinery hydrotreater, hydrocracker and as fuel cells from a H_2-enriched synthetic gas feed. The H_2-enriched synthetic gas feed is usually generated by steam, partial oxidation, or auto-thermal reforming of gas or gasification of solid carbonaceous raw material followed by shift reaction.

Demand for clean fuels is increasing in present society and as such Hydrogen is becoming increasingly important as a clean fuel component that is obtainable from the refining process.

Honeywell UOP have commercialised a cogeneration plant to produce, at the same time, ultrapure hydrogen and power. The Honeywell's process was configured such that the hydrogen PSA off-gas or tail gas is sent to a power island of combined cycle power plants.

Accordingly, it is desirable to provide an improved hydrogen and power generating system and process.

It is also desirable to improve carbon capture from a hydrogen and power generating system.

It is desirable to obtain a capture rate of carbon dioxide in excess of 90%.

SUMMARY OF THE INVENTION

Accordingly, a first aspect of the present invention provides a hydrogen production process operable to produce, at least, hydrogen and carbon dioxide, the process comprises a recycle loop, wherein tail

gas that is produced downstream in the process is used upstream of the process at a shift reactor and for at least one of providing heat for drying coal and providing heat for regenerating CO_2.

The process includes a pressure swing adsorption (PSA) process to produce an ultrapure hydrogen (99.99+%) from the H_2-rich syngas stream. At present the PSA process is a commercially-available process capable of producing ultrapure hydrogen (99.99+%) from the H_2-rich syngas stream.

The process may comprise directing downstream tail gas to an upstream location of the process and providing heat for drying coal and providing heat for regenerating CO_2.

The process may further comprise combusting tail gases at the location of coal drying.

The process may further comprise combusting tail gases at the location of regenerating CO_2.

The process may further comprise feeding air to the location of coal drying and/or the location of regenerating CO_2.

Substantially 100% of tail gas may be reused to provide heat to the upstream units or mixed with a main syngas stream upstream of the process.

A predetermined proportion of tail gas may be captured and reused to provide heat upstream of the process.

A predetermined proportion of tail gas may be transported and mixed with the main syngas stream flowing to the shift reactor.

A predetermined proportion of tail gas may be transported to provide heat to dry coal. A predetermined proportion of tail gas may be transported to provide heat to regenerate CO_2. A further aspect of the present invention provides a hydrogen production plant comprising at least means for drying coal, gasifying means, gas cooling means, means of removing and regenerating carbon dioxide and pressure swing adsorption means operable to separate and exhaust hydrogen from the plant, wherein the plant operates according to the process as claimed in any preceding claim.

The production plant may further comprise means of upstream communication, by which means tail gas is transported to a shift reactor and at least one of the means for drying coal and the means of removing and regenerating carbon dioxide.

The production plant may further comprise means of combusting tail gas proximate the at least one of the means for drying coal and the means of removing and regenerating carbon dioxide and means of imparting heat produced by combusting the tail gas to the at least one of the means for drying coal and the means of removing and regenerating carbon dioxide.

The production plant may further comprise means of proportioning a quantity of tail gas being fed to each of the shift reactor and at least one of the means for drying coal and the means of removing and regenerating carbon dioxide.

Aspects of the invention relate to hydrogen production integrated with carbon capture.

The production plant may comprise carbon capture units, including, but not limited to, a wet absorption, solid-looping fluidised bed process, and fixed-bed adsorption in order to produce CO_2 products of sufficiently high purity for use in, for example, CO_2 storage, the food/beverage industry, enhanced oil/gas recovery and CO_2 utilisation to synthesise valuable chemicals. Gas feed to the H_2 PSA unit normally has an enriched H_2 mole fraction balanced with carbon monoxide (CO), Carbon dioxide (CO_2), Nitrogen (N_2), Argon (Ar), Methane (CH_4) and trace amounts of water vapour and other hydrocarbons.

Adsorption columns of a H_2 PSA process are typically packed with one or more layers of alumina, silica, zeolite, activated carbon and their ion-exchanged forms. The H_2 PSA process operates continuously to produce a product stream of high purity H_2 by selectively adsorbing the impurities during adsorption steps.

Following the adsorption step, each adsorption column experiences one or more stages of depressurising, providing purge, blowdown, purge, one or more stages of repressurising and feed or product pressurisation steps so that the residual hydrogen can be recovered thoroughly and the adsorbents in the column can be regenerated efficiently in order to get the column to recover the adsorption capacity during the adsorption step of the following cycle.

During the blowdown and purge steps gas effluents contain high amounts of CO and CO_2 as well as H_2, N_2, Ar, CH_4 and trace amounts of water vapour and other hydrocarbons. In most cases the H_2 tail gases are utilised as fuel gas since they contain high calorific values. For example, Steam Methane Reforming (SMR) H_2 plants make use of the entire tail gases as fuel gases for endothermic steam reformers.

In a recent publication (UOP, Gasification Technologies 2002) relating to Selexol™, PolySep™ and PolyBed™ operating experience with gasification for power and hydrogen, an advanced process configuration was presented where tail gas is recycled to existing shift reactors to boost the overall H_2 yield in H_2 plants. However, it was identified that increasing the amount of tail gas recycled to shift reactors results in a lower hydrogen mole fraction of the raw H_2 feed due to a build-up of impurities in the recycle loop as indicated in the graph of Figure 1. Figure 1 illustrates the variation of hydrogen mole fraction percentage in the raw H_2 feed with a split ratio of 'tail gas recycle to shift reactors' flow to total tail gas flow. Therefore, such a process configuration would only be possible when a substantial amount of impurities is removed from the recycle loop and where the level of impurities can be maintained at a minimum level such that the downstream H_2 PSA unit can achieve, at the same time,

satisfactory H_2 purity and recovery. Impurities can be reduced, for example by adding a separation process to remove impurities or by bleeding a proportion of tail gas out of the recycle loop.

A process and plant according to aspects of the present invention provide various benefits over existing arrangements. For example, there is lower energy consumption at carbon capture units. This relates to the working capacity at a CO_2 capture unit, wherein by adding thermal energy to the CO_2 regenerator, from the combustion of tail gases, the CO_2 capture plant can be operated with a improved working capacity. Reduced energy consumption and a reduction in size of the equipment in a carbon capture unit is the result.

A further benefit from an increased and more efficient utilisation of the tail gases is increased yield of H_2 in the overall H_2 plant. Generally, part of the synthetic gas leaving a carbon capture unit is being used as a fuel gas for coal drying, which inevitably results in the loss of H_2 product overall. By utilising the tail gas as a fuel gas for coal drying, the loss of the valuable synthetic gas can be avoided. In addition, since the CO_2 capture plant can be operated using less sorbents/solvents because of the improved working capacity, the slip of H_2 into sorbents/solvents at the carbon capture unit can also be reduced.

In addition, the thermal CO_2 regeneration in addition to the depressurised CO_2 regeneration results in reduced power consumption at CO_2 compression. In the event that solvents/sorbents regeneration is done by depressurisation, the heating of the CO_2-laden solvents/sorbents by hot, combusted, tail gas makes it possible to increase CO_2 production at a flash drum operating at an elevated pressure which results in reducing power consumed for CO_2 compression.

Where the aim is to produce high purity CO_2 and ultrapure H_2 with the recoveries of both products as high as 90% in the overall H_2 plant and the H_2 PSA respectively with a tail gas recycle to shift reactors put in place, it is inevitable that part of the tail gas must be bled out or an impurities separator must be utilised because impurities cannot be included in both the pure CO_2 and ultrapure hydrogen products.

The present invention relates to improving the performance of ultrapure hydrogen production plants using dry coal-fed gasification which is integrated with a carbon capture unit with an aim to achieve a capture rate of CO_2 of over 90% and to utilise substantially 100% of tail gases in the production process.

BRIEF DESCRIPTION OF THE DRAWINGS

For a better understanding of the invention and to show how the same may be carried into effect reference will now be made by way of example to the accompanying drawings in which:

Figure 1 is a graphical representation of variation of the hydrogen mole fraction percentage in the raw H_2 feed with the split ratio of the 'tail gas recycle to shift reactors' flow to total tail gas flow;

Figure 2 is a schematic representation of a hydrogen plant using coal gasification with a carbon capture unit in accordance with embodiments of the present invention; and

Figure 3 is a table providing a comparison of the performance of a known H_2 plant and recycle process compared with an H_2 plant and process according to embodiments of the present invention.

DESCRIPTION OF EMBODIMENTS OF THE INVENTION

As discussed above, present technology, which is capable of obtaining a 90% carbon capture rate from a H_2 plant, using coal gasification uses recycled tail gas fed only to shift reactors. However, as illustrated above a problem with this method can be impurities. Therefore, to continue with a process utilising only recycle feed of tail gases to the shift reactors requires additional means to physically remove impurities or means to reduce the quantity of tail gas recycled, by for example bleeding off some tail gas as fuel gases. By such means excessive build-up of the impurities in the recycle loop can be avoided.

Figure 2 illustrates an example system 10 of a H_2 production plant composed of coal gasification, gas conditioning, a carbon capture unit and H_2 Pressure Swing Adsorption (PSA) technology according to embodiments of the present invention where substantially 100% of tail gas is utilised by diverting tail gas upstream to three locations. The illustrated system 10 utilises recycled tail gas at two additional locations compared with the example discussed above.

The production plant comprises carbon capture units such as wet absorption, a solid-looping fluidised bed process, and fixed-bed adsorption in order to produce CO_2 products of sufficiently high purity for use in, for example, CO_2 storage, the food/beverage industry, enhanced oil/gas recovery and CO_2 utilisation to synthesise valuable chemicals. Gas feed to the H_2 PSA unit normally has an enriched H_2 mole fraction balanced with carbon monoxide (CO), Carbon dioxide (CO_2), Nitrogen (N_2), Argon (Ar), Methane (CH_4) and trace amounts of water vapour and other hydrocarbons.

Adsorption columns of a H_2 PSA process are typically packed with one or more layers of alumina, silica, zeolite, activated carbon and their ion-exchanged forms. The H_2 PSA process operates continuously to produce a product stream of high purity H_2 12 by selectively adsorbing the impurities during adsorption steps.

Following the adsorption step, each adsorption column experiences one or more stages of depressurising, providing purge, blowdown, purge, one or more stages of repressurising and feed or product pressurisation steps so that the residual hydrogen can be recovered thoroughly and the adsorbents in the column can be regenerated efficiently in order to get the column to recover the adsorption capacity during the adsorption step of the following cycle.

During blowdown and purge steps, gas effluents contain high amounts of CO and CO_2 as well as H_2, N_2, Ar, CH_4 and trace amounts of water vapour and other hydrocarbons.

According to embodiments of the present invention the H_2 tail gases are utilised upstream to increase the output of ultra pure H_2.

Firstly, a quantity of tail gas 100 is directed upstream to the shift reactor 14 where the tail gas 100 is mixed with the syngas stream flowing from the syngas cooler to improve the hydrogen product yield.

Secondly a quantity of tail gas 200 is directed upstream to the drying gas preparation section 19 where the tail gas 200 is combusted with air 20 to provide heat for drying the coal at the coal dryer 18.

Thirdly, a quantity of the tail gas 300 is directed to a carbon capture unit 22. Again, the tail gas 300 is combusted with air 24 to provide heat, in this case, for CO_2 regeneration.

In the embodiments of the invention substantially 100% of tail gas is recycled with a proportion being fed upstream to the shift reactors 14, a proportion being fed to the drying coal preparation section 19 and a proportion being fed to the CO_2 regenerator 22. According to an embodiment of the present invention, as illustrated in Figure 2, the tail gas is split into three streams 100, 200, 300. One stream 100 is recycled to the shift reactors to boost the H_2 yield at the H_2 PSA. The second stream 200 is sent to 'drying gas preparation' where it is combusted with air to provide the heat for coal drying. The third stream 300 is directed to a CO_2 regenerator of a carbon capture unit, where the tail gas is combusted with air to produce the heat required for more efficient regeneration of CO_2-laden solvents or sorbents. In the illustrated example 21% of tail gas is fed to the shift reactor, 12% of tail gas is sent to drying gas preparation 19 and 67% of tail gas is fed to the CO_2 regenerator.

As explained above, an excessive tail gas recycle to the shift reactors can reduce the H_2 concentration in the raw H_2 stream. As such, the entire quantity of tail gas cannot be recycled to the shift reactors due to the impurities. Therefore, only a proportion of tail gas is fed to the shift reactors; 21% being used in the illustrated example.

The proportion of tail gas recycled to the coal dryer can also be variable because water content in coal can vary with coal type. Therefore, the required amount of tail gas for coal drying will vary depending on the type of coal used in the system and it will be appreciated that it may be possible to use 100% of tail gas for coal drying. It is most likely that a proportion of tail gas will be used. In the illustrated example 12% of tail gas was sent for coal drying purposes.

With further reference to figure 2 and figure 3, the improvement in performance of the H_2 plant according to embodiments of the present invention can be demonstrated. A base case simulation was constructed where the synthetic gas by coal gasification was converted to a H_2-rich stream by shift reaction and subsequently ultrapure hydrogen is produced at the H_2 PSA unit at 286 million standard cubic feet per day (MMSCFD).

Referring to the table illustrated in Figure 3, in Case 1, 21% of tail gas is recycled to upstream of the shift reactor. With the recycle put in place, the raw H_2 feed to the H_2 PSA is as low as 84.5%, which is

lower than 87.1% in the base case (see Figure 3). But the H_2 production rate was improved by 2.4% to 293 MMSCFD from 286 MMSCFD. When recycled, the tail gas is compressed up to the pressure of main syngas stream flowing to the shift reactor, resulting in an increase of total power consumption.

In this example, a conventional dual-stage Selexol™ unit was used to capture Hydrogen Sulphide (H_2S) and CO_2 at the same time. As discussed above, to maintain the H_2 mole fraction in the raw H_2 feed as high as 84.5%, part of the tail gas is removed from (bled out of) the recycle loop and used, for example as fuel gases for other processes. In this example part of the synthetic gas leaving the carbon capture unit is fed to the coal dryer section to provide heat after combustion. This example is referenced as Case 2 in the table of figure 3. In accordance with this embodiment of the present invention, represented as case 2 in figure 3, a portion 200 of the tail gas is utilised for generating hot gases for a coal dryer 18 by combustion while a portion of raw H_2 feed to the H_2 PSA is used as described above. As a result, the ultrapure hydrogen production rate was found to increase by around 1.8% in comparison to Case 1 as indicated in the table of Figure 3.

In accordance with a further embodiment of the invention, represented as case 3 in figure 3, the remainder of the tail gas 300, after its use in the coal dryer, is sent to a dual-stage Selexol™ process. In the dual-stage Selexol™ unit, the CO_2-laden solvents are regenerated by reducing the pressure over two or more flash drums in series. By heating the CO_2-laden solvents flowing to a flash vessel operating at a medium pressure, more CO_2 product can be obtained at the high pressure resulting in improving the solvent working capacity and reducing the power consumption at the CO_2 compression train.

The enhanced solvent working capacity leads to lowering the power consumption for CO_2 capture due to a reduced amount of solvent being pumped and improving the H_2 production rate due to reduced H_2 slip to the circulating solvents.

By utilising substantially 100% of the tail gas at three locations upstream of the process, the production rate of ultrapure hydrogen was found to increase by around 4.2% and the power consumptions at the dual-stage Selexol unit and the CO_2 compression unit was found to reduce by around 15% and 6%, respectively.

Accordingly, it will be appreciated that the total auxiliary power consumption can be reduced by around 8% by the embodiments of the present invention compared with the performances of a system and process where only a limited proportion of the tail gases is recycled for use only with the shift reactors.

While the invention is susceptible to various modifications and alternative forms, specific embodiments are shown by way of example in the drawings and are herein described in detail. It should be understood, however, that drawings and detailed description thereto are not intended to limit the invention to the particular form disclosed.

CLAIMS

1. A hydrogen production process operable to produce, at least, Hydrogen and Carbon dioxide, the process comprises a recycle loop, wherein tail gas produced downstream in the process is used upstream of the process at a shift reactor and at least one of providing heat for drying coal and providing heat for regenerating CO_2.

2. A process as claimed in claim 1, comprising directing downstream tail gas to an upstream location of the process and providing heat for drying coal and providing heat for regenerating CO_2.

3. A process as claimed in any of claims 1 or 2, further comprising combusting tail gases at the location of coal drying.

4. A process as claimed in any preceding claim, further comprising combusting tail gases at the location of regenerating CO_2.

5. A process as claimed in any preceding claim, further comprising feeding air to the location of coal drying and/or the location of regenerating CO_2.

6. A process as claimed in any preceding claim, wherein substantially 100% of tail gas is reused to provide heat to the upstream units or mixed with a main syngas stream upstream of the process.

7. A process as claimed in any preceding claim, wherein a predetermined proportion of tail gas is captured and reused to provide heat upstream of the process.

8. A process as claimed in any preceding claim, wherein a predetermined proportion of tail gas is transported and mixed with a main syngas stream flowing to the shift reactor.

9. A process as claimed in any preceding claim, wherein a predetermined proportion of tail gas is transported to provide heat to dry coal.

10. A process as claimed in any preceding claim, wherein a predetermined proportion of tail gas is transported to provide heat to regenerate CO_2.

11. A hydrogen production plant comprising at least means for drying coal, gasifying means, gas cooling means, means of removing and regenerating carbon dioxide and pressure swing adsorption means operable to separate and exhaust hydrogen from the plant, wherein the plant operates according to the process as claimed in any preceding claim.

12. A hydrogen production plant as claimed in claim 11, further comprising means of upstream communication, by which means tail gas is transported to a shift reactor and at least one of the means for drying coal and the means of removing and regenerating carbon dioxide.

13. A hydrogen production plant as claimed in claim 11 or 12, further comprising means of combusting tail gas proximate the at least one of the means for drying coal and the means of removing and regenerating carbon dioxide and means of imparting heat produced by combusting the tail gas to the at least one of the means for drying coal and the means of removing and regenerating carbon dioxide.

14. A hydrogen production plant as claimed in any of claims 11 to 13, further comprising means of proportioning a quantity of tail gas into proportions being fed to each of the shift reactor and at least one of the means for drying coal and the means of removing and regenerating carbon dioxide.

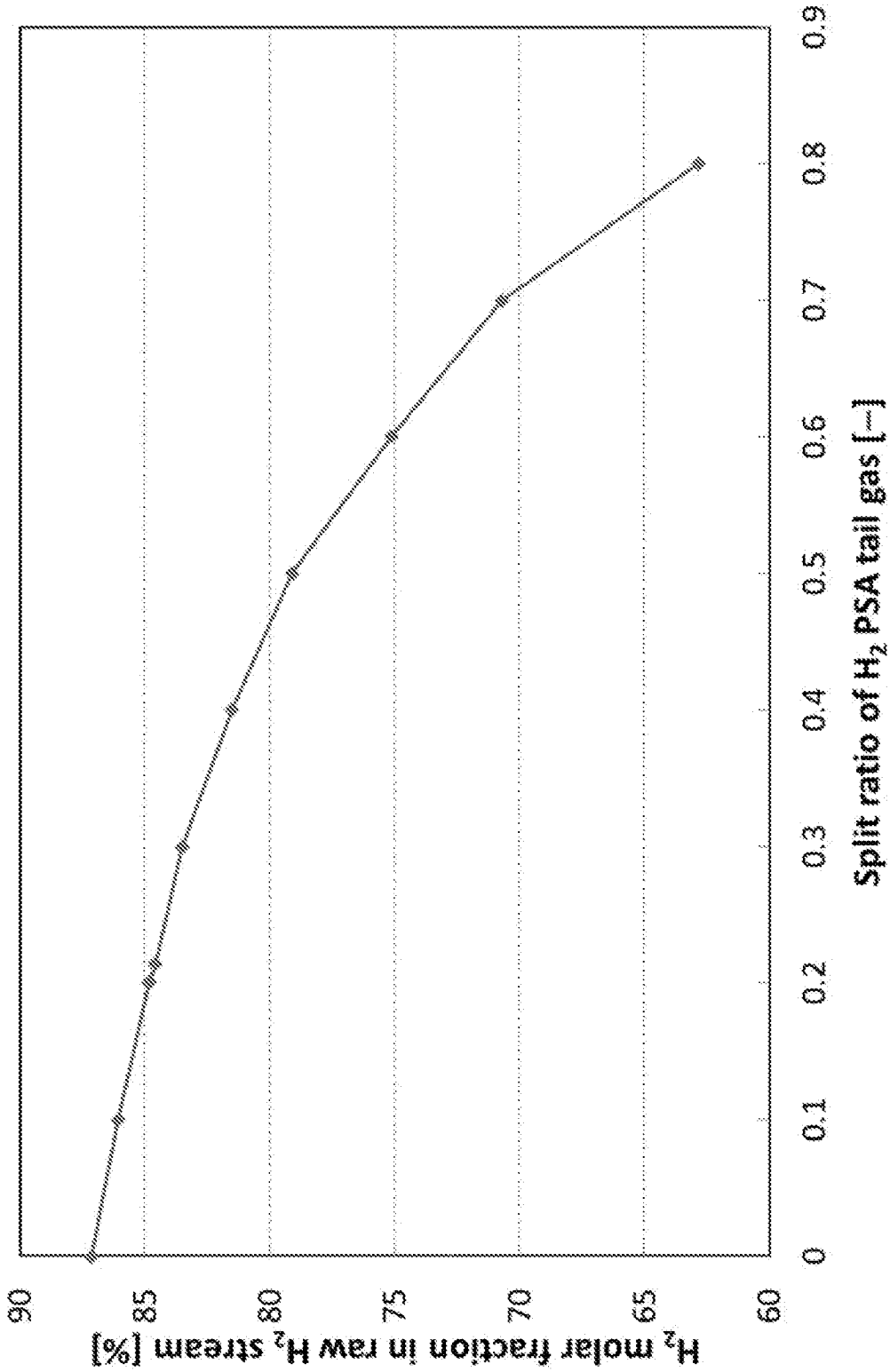
H2 molar fraction in raw H2 stream [%]
90
85
80
75
70
65
60
Split ratio of H2 PSA tail gas [–]
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9

Figure 1

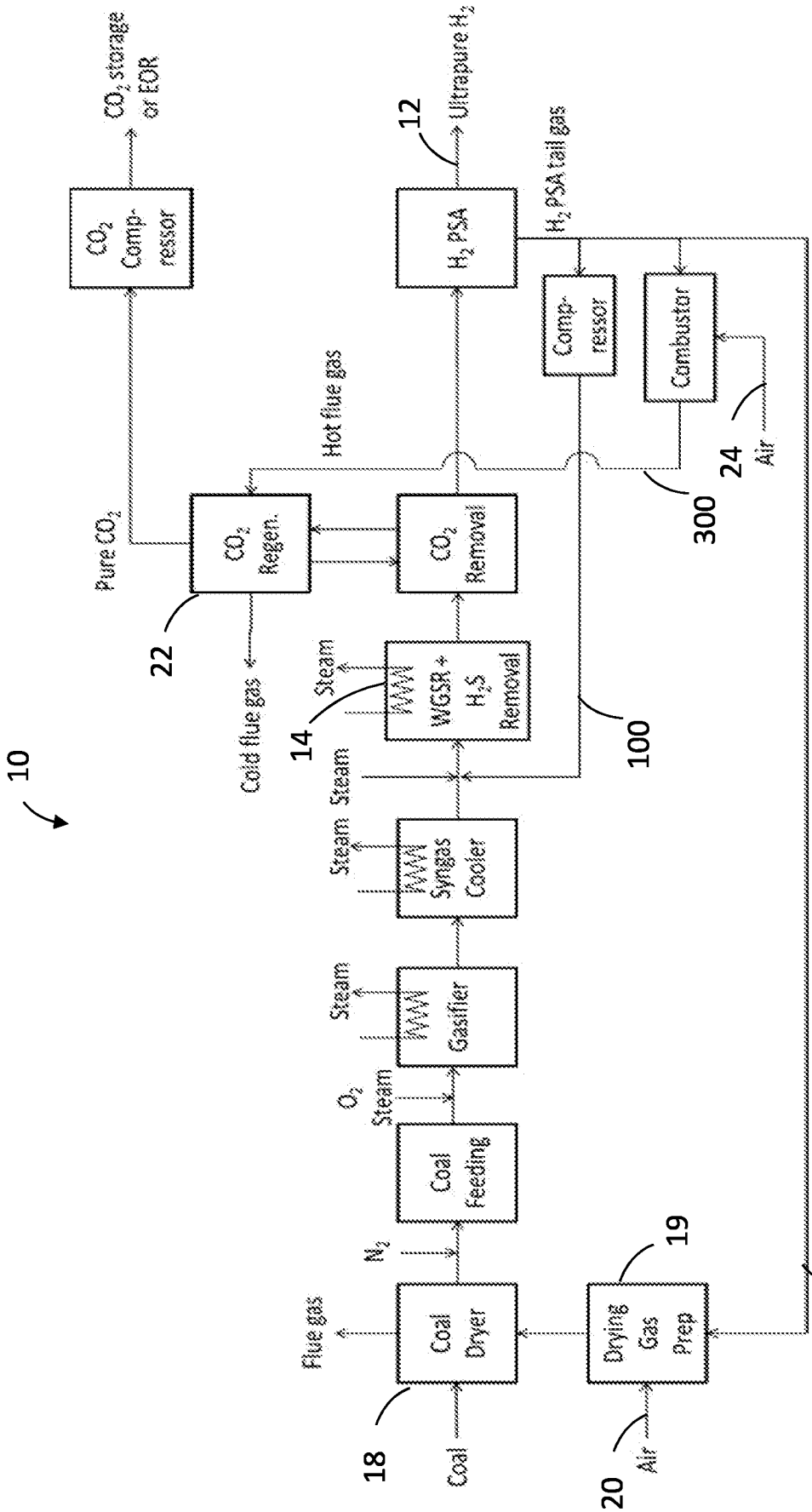
10
CO_2 storage or EOR
CO_2 Comp-ressor
Pure CO_2
CO_2 Regen.
22
Cold flue gas
Hot flue gas
CO_2 Removal
12
Ultrapure H_2
H_2 PSA
H_2 PSA tail gas
Comp-ressor
Combustor
24
Air
300
Steam
14
WGSR + H_2S Removal
100
Steam
Steam
Syngas Cooler
Steam
Gasifier
O_2
Steam
Coal Feeding
N_2
Flue gas
Coal Dryer
19
Drying Gas Prep
200
18
Coal
20
Air

Figure 2

	Base case	*Case 1 (Base case + Tail gas recycle to shift reactors)*	*Case 2 (Case 1 + Tail gas recycle to coal drier)*	*Case 3 (Case 2 + Tail gas recycle to Selexol unit)*
Thermal input [kWt]	1,617,772	1,617,772	1,617,772	1,617,772
Overall CO conversion rate at shift reactors [%]	97.8	97.7	97.7	97.7
Fuel Gas flowrate [MMSCFD]	79	72	62	0
H_2 purity of treated syngas at Selexol unit [%]	87	85	85	85
H_2 recovery of treated syngas at Selexol unit [%]	99.1	99.0	99.0	99.0
H_2S recovery at Selexol unit [%]	99.8	99.8	99.8	99.8
CO_2 recovery at Selexol unit [%]	90.1	90.1	90.1	90.1
CO_2 product purity [%]	97.3	97.1	97.1	97.2
H_2 purity at H_2 PSA [%]	99.99+	99.99+	99.99+	99.99+
H_2 recovery at H_2 PSA [%]	90.0	90.0	90.0	90.0
Ultrapure H_2 production rate [MMSCFD]	**286**	**293 (+2.4%)**	**298 (+4.2%)**	**298 (+4.2%)**
H_2S reboiler duty at Selexol unit [MWt]	14.6	14.6	14.6	11.8 (–19.2%)
Power consumption at Selexol unit [MWe]	**20.2**	**19.3 (–4.5%)**	**19.3 (–4.5%)**	**17.2 (–14.9%)**
Power consumption for CO_2 Compression [MWe]	**31.7**	**31.7**	**31.7**	**29.8 (–6.0%)**
Power consumption at tail gas compressor [MWe]	N/A	3.1	3.1	3.1
Total power consumption [MWe]	**51.9**	**54.1 (+4.4%)**	**54.1 (+4.4%)**	**50.1 (–3.5%)**

Figure 3

INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2014/053843

A. CLASSIFICATION OF SUBJECT MATTER
INV. C10J3/02 C10K1/00 C10K3/04 C01B3/56 B01D53/047
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C10J C10K C01B B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 198 07 224 A1 (LINDE AG [DE]) 26 August 1999 (1999-08-26) figures 1-5 columns 1-4 claims 1-13	1-14
A	WO 2008/157433 A2 (WORMSER ENERGY SOLUTIONS INC [US]; WORMSER ALEX [US]) 24 December 2008 (2008-12-24) figure 8	1-14
A	US 2011/168605 A1 (BLEVINS RANDY [US] ET AL) 14 July 2011 (2011-07-14) figure 1	1-14

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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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Date of the actual completion of the international search	Date of mailing of the international search report
4 March 2015	11/03/2015

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INTERNATIONAL SEARCH REPORT

International application No
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